

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Naoya Ogata et al.

Examiner: Stephen J.Kalafut

Serial No.10/551,330

Group Art Unit: 1795

Filed: September 29, 2005

Confirmation No.1543

Title: COMPOSITE POLYMER ELECTROLYTE COMPOSITION

Honorable commissioner of Patents

and Trademarks, Washington, D. C.

20231

DECLARATION UNDER 37 C.F.R. 1.132.

I, Hiroshi Kagawa, a Japanese citizen residing at 31-102,
Yodono-hara-machi, Takatsuki-city, Osaka-pref, Japan do hereby declare
as follows;

I am a co-inventor in the above-identified application;

I have good working knowledge of the chemistry of electrolyte and
lithium ion battery engineering technology;

Since 2003 to the present date , I have engaged and contributed in
the research,development and production of lithium ion battery to Piotrek
Co., Ltd(former name: Trekion Co., Ltd.).

I have read the Examiner's Office Action in the above-
referenced patent application, mailed on July 29, 2009, and have also
studied Ono et al. JP10-83821, Munshi U S Patent 6,426,863 and Gan
et.6,759,170 relied upon by the Examiner to reject the claims pending in

the application at that time. In my opinion, the invention, as defined by the amended claims, would not have been obvious over the art of record at the time the applicant's invention was made.

In order to demonstrate the difference between Ono et al. Munshi and the claimed invention, the following Comparative experiments were carried out under my supervision.

Comparative experiment 1

Without using a modified polyvinylidene fluoride(DBF) described in Example 2 of our own invention as the Serial No.10/551,330, it has been made a transparent film in 30micron meter thickness under the same conditions of the Experimental Example 3. The film performed ion conductivity at 6.7×10^{-6} S/cm and, and tensile strength at 5 Mpa..

Comparative experiment 2 (Case-1 as no Ionic Liquid formulation)

Without using the composition of quaternary ammonium cation and fluorine-containing anion(so called "Ionic Liquid") described in the Claim 3 of our own invention as the Serial No.10/551,330, it was obtained formerly developed polymer electrolyte consisting of a polyvinylidene fluoride and a copolymer of acrylic monomer or oligomer installing lithium salt. Upon preparing 10 gr of polymer electrolyte consisting of a polyvinylidene fluoride(Kainer#461ex. Atofina as former name of Solvay) and a copolymer of acrylic monomer or oligomer as diacrylic sulphon amide($\text{C}_2\text{H}_2 = \text{C}_2\text{H}_5\text{S}(\text{O})_2\text{NH}_2\text{SO}_2\text{CH}_2 = \text{C}_2\text{H}_2$) copolymerized polyoxyethylene oxide triol, and doping 1 mol LiTFSI (Lithium Bis-Trifluoromethyl-sulphonyl amide) as a kind of lithium salt, it was dissolved in 90g of N-methyl-2-pyrrolidone and dried to make polymer electrolyte film. Under conditions described in Example 7, a layer of cathode electrode/polymer electrolyte /anode electrode and coin cells

installing this layer were made. This cells' capacity were 73% at the initial charge-discharge property and 56% at 20th cycle.

Comparative experiment 3 (Case-2 as no Ionic Liquid formulation)

Without using the composition of quaternary ammonium cation and fluorine-containing anion(so called "Ionic Liquid") described in the Claim 3 of our own invention as the Serial No.10/551,330, it was obtained a polymer electrolyte consisting of polyvinylidene fluoride and acrylic monomer or oligomer_installing lithium salt. Upon preparing 10 gr of polymer electrolyte consisting of a polyvinylidene fluoride(Kainer#461ex. Atofina as former name of Solvay) and acrylic monomer or oligomer as AMPS(2-acrylamido-2-methyl propane sulphonate) described in Munshi's patent (column 5, line 18 ; column 6, lines 3-31; column 11, lines 57-65) and doping 1 mol LiTFSI (Lithium Bis-Trifluoromethyl-sulphonyl amide) as a kind of lithium salt, it was dissolved in 90g of N-methyl-2-pyrrolidone and dried to make polymer electrolyte film. Under conditions described in Example 7, a layer of cathode electrode/polymer electrolyte /anode electrode and coin cells installing this layer were made. This cells' capacity were 67% at the initial charge-discharge property and 48% at 20th cycle.

The results of Comparative experiments 1~3, and the present Examples 3 and 7 are shown in Table A.

Table A

	Property of electrolyte film		Property of coin cell of lithium ion battery	
	Conductivity (S/cm)	Tensile strength (Mpa)	Discharge capacity at the initial cycle (%)	Discharge capacity at the 20th cycle (%)
Comparative experiment 1	6.7×10^{-6}	5	—	—
The present Example 3	2.1×10^{-3}	11	—	—
Comparative experiment 2	—	—	7 3	5 6
Comparative experiment 3	—	—	6 7	4 8
The present example 7	—	—	9 5	8 5

Low temperature property :Comparative experiment - 2 possibly operated 50% capacity at -10°C Comparative experiment - 3 possibly operated 35% capacity at -10°C The present Example 7 steadily operating 70% capacity at -10°C Discussion

The results of Comparative experiment 1, set forth in Table A, show that Comparative experiment 1 is inferior to the present Example 3 in conductivity and tensile strength. The results of Comparative experiments 2 and 3 set forth in Table A, show that Comparative experiments 2 and 3

are much inferior the capacity and cycle property to the present Example 7 in conductivity at the 20th cycle, of which reasons are as follows; In particular, the results of Comparative experiment 2 indicates that the polymer electrolyte influences to deteriorate remarkably the capacity performance because of increasing much impedance as the internal resistance on the surface of both electrodes, in particular, by deriving lithium sulphonate salts to become generation of a dendolite as one of critical reasons deteriorating remarkably the cycle property(REDOX). In addition, those sulphon or sulphonate derivatives have been generally evaluated as a restraining characteristics of forming SEI(surface electrolyte intermediate) on surface of anode electrode layer, e.g. a graphite active material.

This disadvantage of Comparative experiment 3 disclosed by Munshi(US 6,426,863) can be made a solution by execution through the Claim 3 of our own invention as the Serial No.10/551,330 to decrease remarkably such impedance by formulating the composition of the Ionic Liquid.

Our original technology utilized acrylic monomer or oligomer such as AMPS (Comparative experiment 3) and acrylic monomer or oligomer for making PEO triol copolymer and acrylic monomer or oligomer precursor (Comparative experiment 2) promoted since 1984 ,and be judged to abandon this technology in 1999 and newly promoted the polymer electrolyte technology in our own invention as the Serial No.10/551,330 in polymer electrolyte development history record aspect

In conclusion, the results of Comparative experiment 1 set forth in Table A show that it is inferior to the present Example 3 in conductivity and tensile strength. Also, the results of Comparative experiments 2 and 3 set forth in

Table A, show that Comparative experiments 2 is much inferior to the present Example 7 in conductivity at the 20th cycle.

I therefore do not believe that the improvement and substantial use level would have been expected from Ono et al. and Munshi.

I, the undersigned petitioner, declare further that all statements made herein are true and that all statements made on information and belief are believed to be true; and

further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

September 14, 2009



Hiroshi Kagawa